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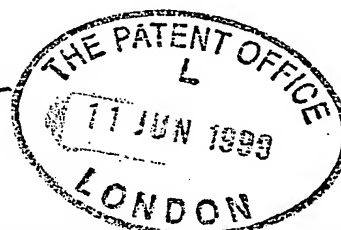
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Request for grant of a patent

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1. Your reference	91341/PRS/MNE		
2. Patent application number (The Patent Office will fill in this part)	9913695.4		
3. Full name, address and postcode of the or of each applicant (underline all surnames)	CAMBRIDGE DISPLAY TECHNOLOGY LIMITED 181A Huntingdon Road Cambridge CB3 0DJ		
Patents ADP number (if you know it)	G166.441002		
If the applicant is a corporate body, give the country/state of its incorporation	United Kingdom		
4. Title of the invention	Method of Producing Organic Light-Emissive Devices		
5. Name of your agent (if you have one)	Page White & Farrer		
"Address for service" in the United Kingdom to which all correspondence should be sent (including the postcode)	54 Doughty Street London WC1N 2LS		
Patents ADP number (if you know it)	1255003		
6. If you are declaring priority from one or more earlier patent applications, give the country and the date of filing of the or of each of these earlier applications and (if you know it) the or each application number	Country	Priority application number (if you know it)	Date of filing (day / month / year)
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Description 15

Claim(s) 5

Abstract

Drawing(s) 4 + 4



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Priority documents

Translations of priority documents

Statement of inventorship and right to grant of a patent (Patents Form 7/77)

Request for preliminary examination and search (Patents Form 9/77) 1

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11. I/We request the grant of a patent on the basis of this application.

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PAGE WHITE & FARRER

12. Name and daytime telephone number of person to contact in the United Kingdom P.R. Slingsby 0171-831-7929

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DUPLICATE

METHOD OF PRODUCING ORGANIC LIGHT-EMISSIVE DEVICES

The present invention relates to a method for producing optoelectronic devices, in particular, organic light-emissive devices (OLEDs).

Organic light-emissive devices typically comprise an organic light-emissive region sandwiched between two electrodes such that charge carriers can move between the organic light-emissive region and the two electrodes.

In many applications such as active matrix displays, at least one of the electrodes and the light-emissive region is required to be patterned into an ordered array of discrete pixels whereby each pixel can be addressed separately. Full colour active matrix displays require a light-emissive region comprising discrete pixels of three different light-emissive materials which are respectively capable of emitting light in the three primary colour regions.

One method of providing a patterned organic light-emissive region involves the use of ink-jet printing. However, the degree of pixel density which can be achieved by ink-jet printing is severely limited. Furthermore, the droplets formed by ink-jet printing are hemispherically shaped leading to non-uniform illumination within each pixel. In addition, it is an expensive process particularly when used to produce large area displays.

According to a first aspect of the present invention, there is provided a method for forming a patterned layer of a light-emissive material on a substrate, comprising

the steps of: providing a holed layer on the surface of the substrate, the holed layer defining a plurality of holes through which the underlying substrate is exposed; and applying a light-emissive material to the surface of the holed layer opposite the substrate and displacing the light-emissive material in fluid form across the surface of the holed layer whereby it is deposited in the holes of the holed layer.

The method of the present invention can be used to form any patterned layer such as a series of parallel rows or columns, a two-dimensional array of discrete pixels, or a layer in the shape of a legend comprising, for example, one or more letters or numerals or other shapes.

The light-emissive material is preferably displaced across the surface of the holed layer such that it is selectively deposited in the holes of the holed layer, i.e. such that it is deposited in the holes of the holed layer without substantially any deposition of the light-emissive material on the surface of the holed layer.

In a preferred embodiment, the holed layer is permanently adhered to the substrate. In this embodiment, the holed layer is not intended to be subsequently removed from the substrate.

The light-emissive material is preferably an organic light-emissive material. The term organic light-emissive material includes precursors of organic light-emissive materials, the precursors not necessarily being light-emissive themselves. For example, the organic light-emissive material may be a semi-conductive conjugated

polymer or a precursor of a semi-conductive conjugated polymer.

The holed layer is preferably made of a polymer which is electrically non-conductive, and does not chemically react with the light-emissive material in a way which renders the light-emissive material non light-emissive. The holed layer may be opaque or transparent.

The holed layer is preferably made of a relatively benign polymer such as a fluorinated polymer. Examples of suitable fluorinated polymers include amorphous polytetrafluoroethylene (PTFE) or an amorphous ethylene-tetrafluoroethylene copolymer (ETFE), with amorphous PTFE being particularly suitable. The use of fluorinated polymers is advantageous because they do not react and swell upon contact with the standard solvents such as xylene and water which are typically used for forming solutions of organic light-emissive materials.

The holed layer is preferably formed by providing a layer of a polymer on the substrate, and then forming a plurality of holes in the polymer layer by etching. The etching is preferably carried out by a reactive ion etching process using an oxygen-based plasma. The polymer layer may be provided on the substrate by deposition using a standard technique such as spin-coating or blade-coating, or by laminating a pre-formed polymer layer to the substrate.

Alternatively, a polymer layer with pre-formed holes could be prepared in advance and then laminated to the substrate.

The thickness of the holed layer will clearly depend on the desired thickness of the patterned layer of light-emissive material, since its thickness will determine the thickness of the patterned layer of light-emissive material.

The light-emissive material is preferably applied to the surface of the holed layer as a solution thereof in a suitable solvent. Insoluble materials can be applied, for example, as a suspension thereof in a suitable liquid medium or in molten form.

According to a second aspect of the present invention, there is provided a method for forming an optoelectronic device comprising the steps of: providing a substrate having a patterned first electrode on a surface thereof; providing a holed layer on the surface of the substrate having the patterned first electrode, the holed layer defining a plurality of holes through which the patterned first electrode is exposed; applying a light-emissive material to the surface of the holed layer opposite the substrate, and displacing the light-emissive material in fluid form across the surface of the holed layer whereby it is deposited into the holes of the holed layer; solidifying the light-emissive material; and forming a second electrode on each region of solid light-emissive material in the holes such that charge carriers can move between the light-emissive material and the second electrode.

The light-emissive material can be deposited directly on the underlying patterned first electrode. Alternatively, intermediate layers such as charge transport layers or additional light-emissive layers can be provided between

the patterned first electrode and the light-emissive material provided that they allow the movement of charge carriers between the patterned first electrode and the light-emissive material. In a preferred embodiment, a layer of a hole-injection material is provided between the patterned first electrode and the light-emissive layer to promote the injection of holes from the patterned first electrode into the light-emissive material.

Likewise, the second electrode can be formed directly on the layer of light-emissive material, or intermediate layers such as charge transport layers or additional layers of light-emissive material can be interposed between the layer of light-emissive material and the second electrode, provided that they allow the movement of charge carriers between the second electrode and the layer of light-emissive material.

The light-emissive material is preferably an organic light-emissive material such as a semi-conductive conjugated polymer. The term conjugated polymer includes polymers and oligomers which are conjugated along the entire length thereof, and polymers and oligomers which comprise conjugated segments separated by non-conjugated segments.

The holed layer is preferably formed by depositing a layer of a polymer on the substrate and then forming the plurality of holes in the polymer layer after deposition of the holed layer. The plurality of holes are preferably formed by etching of the polymer layer, such as anisotropic reactive ion etching.

A patterned array of one or more holes of the desired shape may be achieved by a photolithographic technique. Such a technique involves forming a layer of a photoresist on the surface of the polymer layer; exposing the whole area of the photoresist (for example, using a shadow mask) to radiation; removing portions of the photoresist layer by the use of a suitable developer to leave a photoresist layer having one or more holes of the desired shape formed therein which expose the portions of the underlying polymer layer in which holes are to be formed. Subsequent exposure to an etching plasma will only etch the portions of the polymer layer left exposed by the holes in the photoresist layer.

The etching is preferably carried out using a plasma which not only serves to etch the material of the holed layer but also serves to treat and prepare the exposed surface of the patterned first electrode for deposition of subsequent layers.

Embodiments of the present invention will be described hereunder, by way of example only, with reference to the accompanying drawings, in which:-

Figures 1(a) to 1(h) are schematical cross-sectional views of a single colour organic light-emissive device at various stages of its production according to a first embodiment of the present invention.

Figures 2(a) to 2(h) are schematical cross-sectional views of a full colour organic light-emissive device at

various stages of its production according to a second embodiment of the present invention.

First, with reference to Figures 1(a) to 1(h), a method of producing an organic light-emissive device according to a first embodiment of the present invention will be described.

A layer of a fluorinated polymer 6 such as amorphous polytetrafluoroethylene (PTFE) is formed over the surface of a substrate comprising a glass base 2 having a two-dimensional ordered array of anode pixels 4 made of indium tin oxide (ITO) formed on a surface thereof. In this embodiment, the substrate is made of glass, but it can also be made of other materials such as plastics eg. PMMA. Wiring (which is not shown in the Figures) is connected from the periphery of the glass base to each anode pixel so that each anode pixel can be separately addressed in the final device.

The two-dimensional array of anode pixels 4 can be formed on a glass base 2 according to a conventional method such as sputtering using a shadow mask. The layer of fluorinated polymer is formed by a conventional coating method such as spin-coating using a solution of the fluorinated polymer in a suitable organic solvent and then drying.

The thickness of the layer of the fluorinated polymer layer will determine the thickness of the organic light-emissive region in the final device. For a typical organic light-emissive device, the fluorinated polymer layer is deposited to a thickness in the range of 50 to 100nm.

Holes 8 are then created in the fluorinated polymer layer 6 to expose portions of the underlying anode pixels 4. The holes 8 are created by the following process. The surface of the fluorinated polymer layer is provided with an appropriately patterned photopolymer layer such as a photoresist on its upper surface by a standard photolithographical technique. The use of projection lithography to produce the patterned photopolymer layer can give affordable and high throughput processing.

Etching of the regions of the fluorinated polymer layer 6 left exposed by the photoresist is then carried out by a plasma etching process using an oxygen-based chemistry to form vertical holes in the fluorinated polymer layer 6 which extend down to the surface of the anode pixels 8. Each anode pixel 4 serves as an etch-stop. Once etching is completed the photoresist is stripped from the surface of the holed layer 6. The oxygen plasma also serves to treat the surface of the ITO anode pixels 4 for subsequent deposition of organic layers.

Although an oxygen plasma is used in this embodiment, other gases such as freons and sulphur hexafluoride can be used to carry out the etching process, with or without dilutions of oxygen. Alternatively, argon could be used where selectivity is not a requirement.

The exposed surface of the ITO anode pixels is then cleaned. Once the surface is cleaned, another short oxygen plasma treatment may be required to recover the treated ITO surface.

Next, a solution 12 of polystyrene sulphonic acid doped polyethylene dioxythiophene ("PEDOT-PSS") in water is applied to the surface of the holed polymer layer 6 and is wiped across the surface of the holed polymer layer 6 using a push rod 10, whereby the PEDOT-PSS solution 12 becomes deposited in the holes 8 to completely fill the holes 8 without substantially any deposition of the PEDOT-PSS solution on the top surface of the holed layer 6. The PEDOT-PSS solution 12 in the holes 8 is then dried to leave PEDOT-PSS pixels 14 in the holes 8. The PEDOT-PSS pixels only partially fill the holes 8 as a consequence of the reduction in volume associated with the evaporation of the aqueous solvent.

The thickness of the PEDOT-PSS pixels can be controlled by adjusting the concentration of the PEDOT-PSS solution. The thickness of the PEDOT-PSS pixels may be reduced by using a more dilute solution, and may be increased by using a more concentrated solution.

The use of a fluorinated polymer for the holed layer 6 is advantageous in this embodiment because it does not react and swell upon contact with the aqueous solvent.

The PEDOT-PSS provides a hole-injection layer which promotes injection of positive charge carriers into the light-emissive material to be subsequently deposited. Other materials which provide this hole-injecting function can also be used such as poly(2,7-(9,9-di-n-octylfluorene)-(1,4-phenylene-(4-imino(benzoic acid))-1,4-phenylene-(4-imino(benzoic acid))-1,4-phenylene)) ("BFA"), polyaniline and poly(p-phenylenevinylene) ("PPV").

Next, a solution 16 of an organic light-emissive material such as a semi-conductive conjugated polymer is applied to the surface of the holed layer 6 and is wiped across the surface of the holed layer 6 using a push rod 10 whereby it is selectively deposited in the holes 8 in the holed layer 6 on top of the PEDOT-PSS pixels 14. The solution of the organic light-emissive material 16 left deposited in the holes 8 of the holed layer 6 is then dried to leave each hole containing an underlying layer 14 of PEDOT-PSS and an overlying layer 18 of the organic light-emissive material.

The organic light-emissive material may comprise one or more individual organic materials, suitably polymers, preferably fully or partially conjugated polymers. Suitable materials include one or more of the following in any combination: poly(p-phenylenevinylene) ("PPV"), poly(2-methoxy-5(2'-ethyl)hexyloxyphenylenevinylene) ("MEH-PPV"), one or more PPV-derivatives (e.g. di-alkoxy or di-alkyl derivatives), polyfluorenes and/or co-polymers incorporating polyfluorene segments, PPVs and related co-polymers, poly(2,7-(9,9-di-n-octylfluorene)-(1,4-phenylene-((4-secbutylphenyl)imino)-1,4-phenylene)) ("TFB"), poly(2,7-(9,9-di-n-octylfluorene) - (1,4-phenylene-((4-methylphenyl)imino)-1,4-phenylene-((4-methylphenyl)imino) - 1,4-phenylene)) ("PFM"), poly(2,7-(9,9-di-n-octylfluorene) - (1,4-phenylene-((4-methoxyphenyl)imino)-1,4-phenylene-((4-methoxyphenyl)imino)-1,4-phenylene)) ("PFMO"), poly(2,7-(9,9-di-n-octylfluorene) ("F8") or (2,7-(9,9-di-n-octylfluorene)-3,6-Benzothiadiazole) ("F8BT"). Alternative materials include small molecule materials such as Alq3.

A typical solvent for forming solutions of the above materials is xylene. Other suitable solvents are THF, trichloromethane, toluene and other aromatic solvents. When these are used as the solvent, the use of a fluorinated polymer for the holed layer is advantageous because it does not swell upon contact with them.

A cathode 20 is then deposited over the surface of the light-emissive pixels 18 and the surface of the holed layer 6. The cathode preferably has a double layer construction with a thin underlying layer of a reactive metal such as calcium deposited on top of the light-emissive pixels 18 and a thick layer of a non-reactive metal such as aluminium deposited on top of the thin layer of reactive metal. The cathode is preferably deposited by evaporation to reduce to a minimum any damage to the underlying organic layers.

The matrix of polymer material surrounding each organic pixel and each anode pixel remains in the final device, and further serves to both electrically insulate the organic pixels and anode pixels from the influence of adjacent organic pixels and anode pixels. It also serves to physically protect and give structural support to the organic pixels and anode pixels.

Next, with reference to Figures 2(a) to 2(h), a method of producing a full colour organic light-emitting device according to a second embodiment of the present invention will be described.

A layer 54 of a fluorinated polymer is formed on a substrate comprising a glass base 50 having a two-dimensional array of anode pixels 52 made of ITO

deposited thereon. The fluorinated polymer layer 54 may be formed, for example, by the method described for the first embodiment.

A first set of holes 56 are then formed at selected points of the fluorinated polymer layer 54 in the same way as in the first embodiment, to expose only some of the anode pixels 52. For example, the two dimensional array of anode pixels may comprise an array of groups of three associated pixels, with the first set of holes 56 being formed to expose only a single one of the three anode pixels in each group.

A layer of an organic hole-injection material and a layer of a first organic light-emissive material are then deposited in each hole 56 in the same manner as in the first embodiment to create an array of organic pixels 58. A cathode 60 is then formed over the whole surface of the holed layer 52 and the organic pixels 58. As in the first embodiment, the cathode preferably has a double layer construction with an underlying first thin layer of a reactive metal such as calcium and an overlying thick layer of a non-reactive metal such as aluminium.

The cathode metallization serves to protect the organic pixels 58 embedded into the fluorinated polymer from subsequent processing steps.

Next, a second set of holes 62 is formed to expose some of the anode pixels 52 which were not exposed in the previous hole-forming step. For example, in the array described above, the second set of holes could be formed to only expose a second one of the three anode pixels in each group. This second set of holes 62 can be formed in

the same manner as the first set of holes, except that different plasma chemistries may be needed, one to punch through the cathode 60 and one to etch through the fluorinated polymer layer 54. Fluorocarbons, sulphur hexafluoride, argon, methane/hydrogen and chlorine chemistries such as boron trichloride are examples of materials which can be used to punch through the cathode. The etching through of the cathode can be observed by optical absorption in the plasma chamber used. The etching of the fluorinated polymer layer 54 is carried out using an oxygen-based plasma as in the first embodiment.

A second set of organic pixels 64 is then formed in the second set of holes 62 in the same manner as in the first embodiment. Although, not shown in the Figures, each of the second set of organic pixels 64 also comprises an underlying layer of PEDOT-PSS (as a hole injection layer) and an overlying layer of an organic light-emissive organic material which is capable of emitting light of a primary colour different to that of the light-emissive material deposited in the first set of holes 56.

Another cathode 66 is then deposited over the second set of organic pixels 64 to protect the second set of organic pixels 58 from subsequent processing steps.

Next, a third set of holes 68 is formed through the cathodes 60,66 and the fluorinated polymer layer 54 to expose the remaining anode pixels which were not exposed in the previous first and second hole forming steps. For example, in the array described above, the third set of holes could be formed to only expose the third and final anode pixel of each group of three associated anode

pixels. The third set of holes 68 can be formed in the same manner as the second set of holes 62. A third set of organic pixels 70 is then deposited in the third set of holes 68 in the same manner as in the first embodiment. Although not shown in the Figures, the third set of organic pixels 68 also each comprise an underlying layer of PEDOT-PSS as a hole-injection layer and an overlying layer of an organic light-emissive material which is capable of emitting light of a primary colour different from the light-emissive materials deposited in the first and second sets of holes 56, 62.

In both the above-described embodiments, the fact that the shape and size of the organic pixels are defined by holes which are created by etching means that pixels of uniform shapes and of very small dimensions (of the order of a number of microns) can be formed because of the high precision of the etching process, and the pixel density can therefore be increased significantly compared to arrays of organic pixels produced by conventional methods such as ink-jet printing.

Furthermore, this method can be used to produce pixels having a uniform rectangular shape i.e. having a depth greater than their length or width. This gives good colour uniformity across the individual pixel area.

Furthermore, the above-described method of deposition of the organic materials is an extremely cost-effective deposition method which is particularly significant in the production of large area displays.

Although the present invention has been described above in respect of its application to the formation of an

organic layer comprising an array of pixels, it will be appreciated by the person skilled in the art that the method of the present invention can be used to form other patterned layers such as layers comprising one or more parallel rows or columns, each column or row comprising the same light-emissive material or comprising light-emissive materials which emit light of different colours. Alternatively, the method of the present invention can be used to form layers in the shape of one or more legends such as ones comprising one or more numerals or letters or other shapes, each numeral, letter or other shape composing the legend comprising the same light-emissive material or comprising light-emissive materials which emit light of different colours.

CLAIMS

1. A method for forming a patterned layer of a light-emissive material on a substrate, comprising the steps of: providing a holed layer on the surface of the substrate, the holed layer defining a plurality of holes through which the underlying substrate is exposed; and applying a light-emissive material to the surface of the holed layer opposite the substrate and displacing the light-emissive material in fluid form across the surface of the holed layer whereby it is deposited in the holes of the holed layer.
2. A method according to claim 1 wherein the light-emissive material is selectively deposited in the holes of the holed layer.
3. A method according to claim 1 or claim 2 wherein the holed layer is permanently adhered to the substrate.
4. A method according to any of claims 1 to 3 wherein the holed layer is provided by laminating a polymer layer with pre-formed holes to the surface of the substrate.
5. A method according to any of claims 1 to 3 wherein the holed layer is provided by providing a layer of a polymer on the substrate, and then forming the plurality of holes in the polymer layer.
6. A method according to claim 5 wherein the holes are formed in the polymer layer by etching.
7. A method according to claim 6 wherein the etching is carried out using an oxygen-based plasma.
8. A method according to claim 6 or 7 wherein the etching is carried out by a plasma etching process.

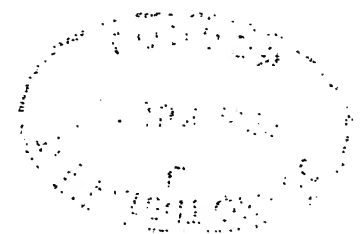
9. A method according to claim 5 wherein the polymer layer is provided by coating the substrate with the polymer.
10. A method according to claim 5 wherein the polymer layer is provided by laminating a pre-formed polymer layer to the substrate.
11. A method according to any preceding claim wherein the holed layer comprises an electrically insulating polymer.
12. A method according to claim 11 wherein the electrically insulating polymer is a fluorinated polymer.
13. A method according to any preceding claim wherein the thickness of the holed layer is in the range of 50 to 100nm.
14. A method for forming an optoelectronic device comprising the steps of:
 - providing a substrate having a patterned first electrode on a surface thereof;
 - providing a holed layer on the surface of the substrate having the first patterned electrode, the holed layer defining a first set of holes through which the patterned first electrode is exposed;
 - applying a first light-emissive material to the surface of the holed layer opposite the substrate, and displacing the first light-emissive material in fluid form across the surface of the holed layer whereby it is deposited into the first set of holes of the holed layer;
 - solidifying the first light-emissive material; and
 - forming a second electrode on the solidified first light-emissive material in the first set of holes such that charge carriers can move between the first light-emissive material and the second electrode.

15. A method for forming an optoelectronic device according to claim 14 wherein the first light-emissive material is selectively deposited into the first set of holes.
16. A method for forming an optoelectronic device according to claim 14 or claim 15 wherein the holed layer is permanently adhered to the substrate.
17. A method for forming an optoelectronic device according to any of claims 14 to 16 wherein the holed layer is provided by laminating a polymer layer with pre-formed holes to the surface of the substrate.
18. A method for forming an optoelectronic device according to any of claims 14 to 16 wherein the holed layer is provided by providing a layer of a polymer on the surface of the substrate having the patterned first electrode, and then forming the first set of holes in the polymer layer.
19. A method for forming an optoelectronic device according to claim 18 wherein the first set of holes are formed by etching.
20. A method for forming an optoelectronic device according to any one of claims 14 to 19 wherein the patterned first electrode comprises a two-dimensional array of discrete pixel electrodes.
21. A method according to claim 20 wherein the first set of holes only expose some of the discrete pixel electrodes.
22. A method according to claim 14 further comprising forming a second set of holes in the holed layer to expose discrete pixel electrodes which were not exposed by the first set of holes in the holed layer, depositing a layer of second light-emissive material in the second set of holes and forming a

third electrode on the layers of second light-emissive material such that charge carriers can move between the layers of second light-emissive material and the third electrode.

23. A method according to claim 22 wherein the second electrode is additionally formed over substantially the entire surface of the holed layer opposite the substrate, and the second set of holes is formed by etching through the second electrode and the holed layer.
24. A method according to claim 23 further comprising forming a third set of holes in the holed layer to expose further discrete pixel electrodes which were not exposed by the first or second set of holes, depositing a layer of third light-emissive material in the third set of holes, and forming a fourth electrode on the layers of third light-emissive material such that charge carriers can move between the fourth electrode and the layers of third light-emissive material.
25. A method according to claim 24 wherein the first, second and third light-emissive materials are each capable of emitting light of the three primary colours respectively.
26. A method according to any of claims 14 to 25 further comprising the step of forming a layer of a charge transport material in the first set of holes.
27. A method according to claim 26 wherein the layer of charge transport material is deposited in the first set of holes by applying the charge transport material to the surface of the holed layer and displacing the charge transport material in a fluid form across the surface of the holed layer whereby it is deposited in the first set of holes.

28. A method for forming an optoelectronic device according to any one of claims 14 to 27 wherein the second electrode is deposited without first removing the holed layer.
29. A method for forming an optoelectronic device according to claim 24 wherein the second, third and fourth electrodes are formed without removing the holed layer.
30. An optoelectronic device produced by the method of any of claims 14 to 29.
31. An optoelectronic device according to claim 30 wherein the holed layer remains in the final device.
32. A method of forming a patterned layer of a light-emissive material on a substrate substantially hereinbefore described with reference to Figures 1(a) to 1(h) or Figures 2(a) to 2(h) of the accompanying drawings.
33. A method for forming an optoelectronic device substantially as hereinbefore described with reference to Figures 1(a) to 1(h) or Figures 2(a) to 2(h) of the accompanying drawings.
34. An optoelectronic device produced by the method substantially as hereinbefore described with reference to Figures 1(a) to 1(h) or Figures 2(a) to 2(h) of the accompanying drawings.



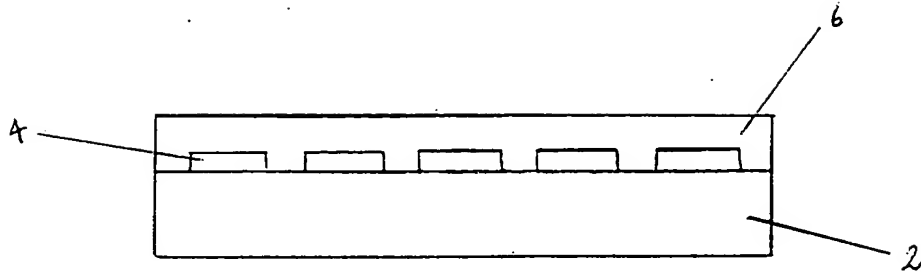


Fig. 1(a)

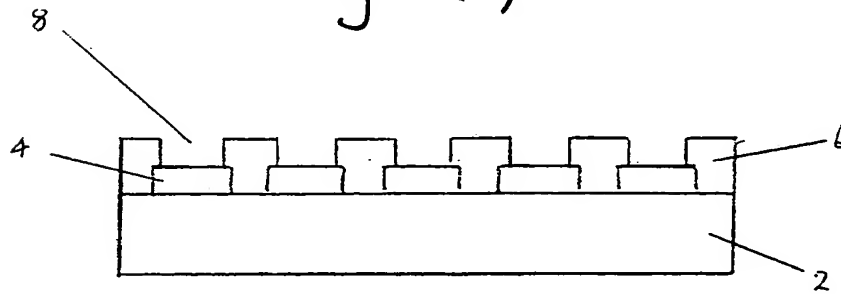


Fig. 1(b)

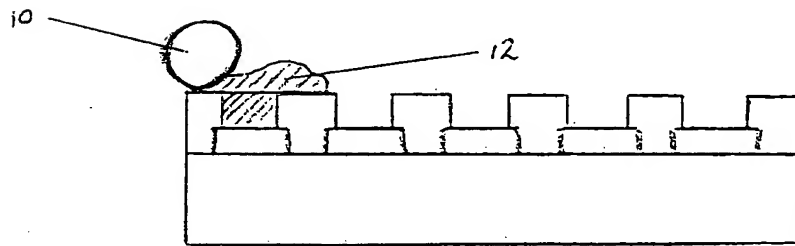


Fig. 1(c)

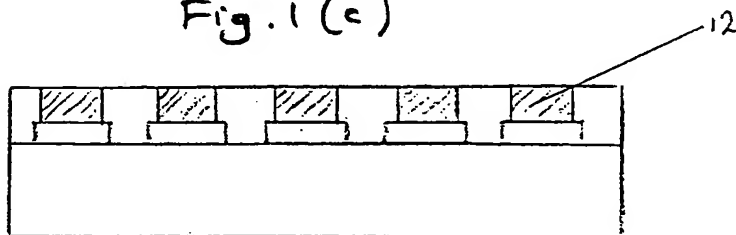


Fig. 1(d)

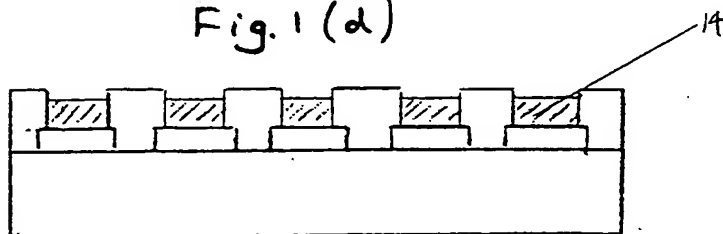


Fig. 1(e)

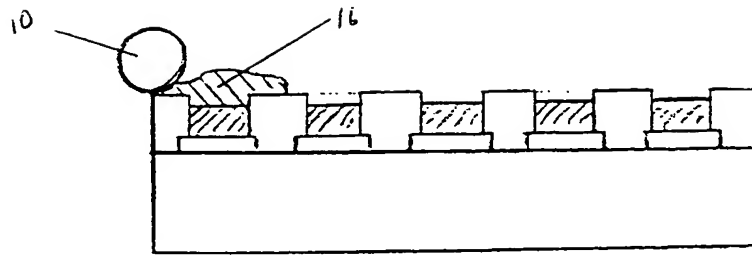


Fig. 1(f)

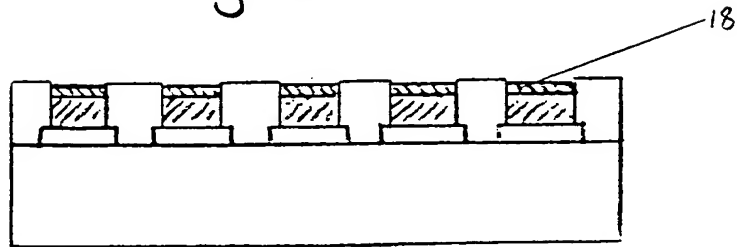


Fig. 1(g)

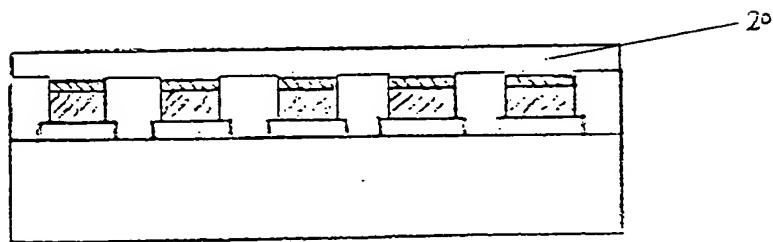


Fig. 1(h)

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NOT TO BE REPRODUCED

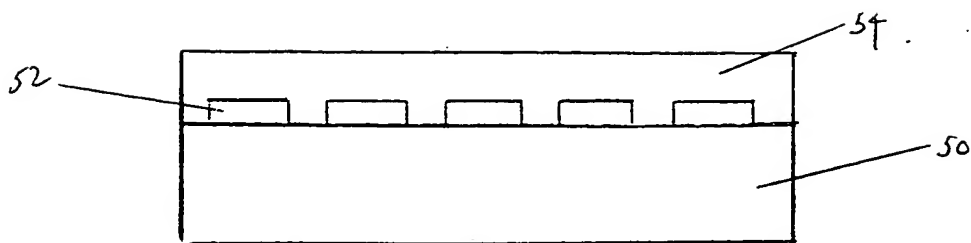


Fig. 2(a)

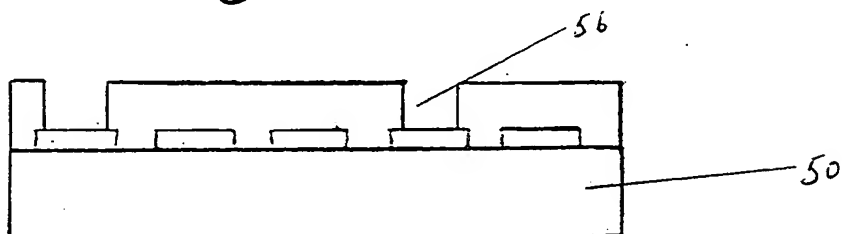


Fig. 2(b)

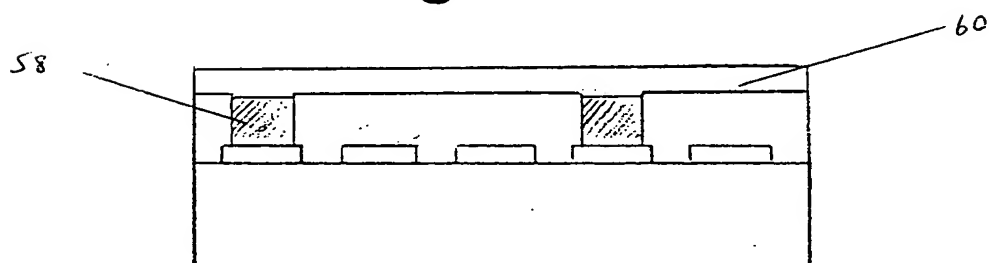


Fig. 2(c)

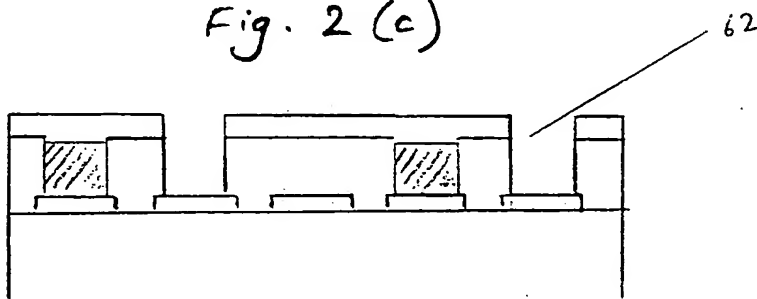


Fig. 2(d)

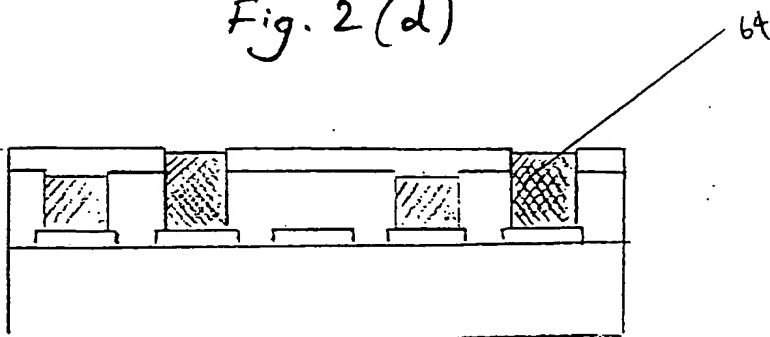


Fig. 2(e)

4(4)

NOT TO BE REPRODUCED

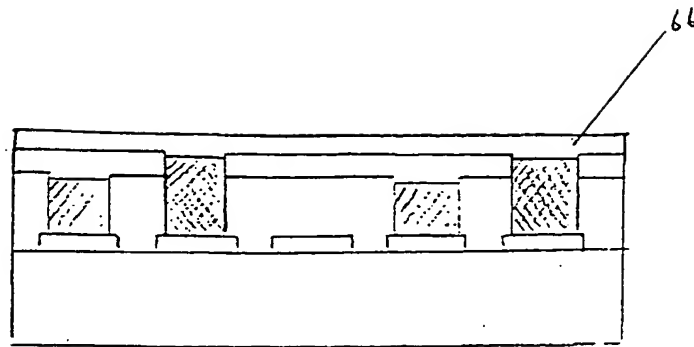


Fig. 2(f)

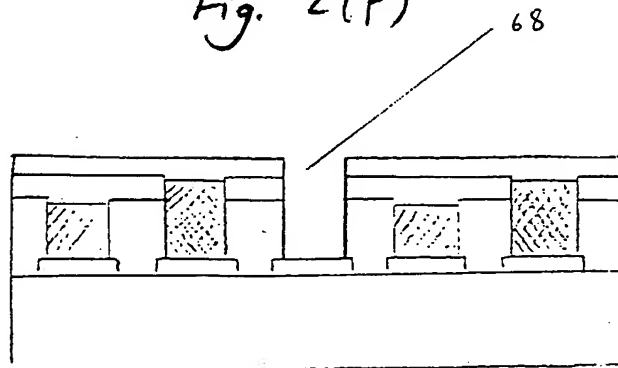


Fig. 2(g)

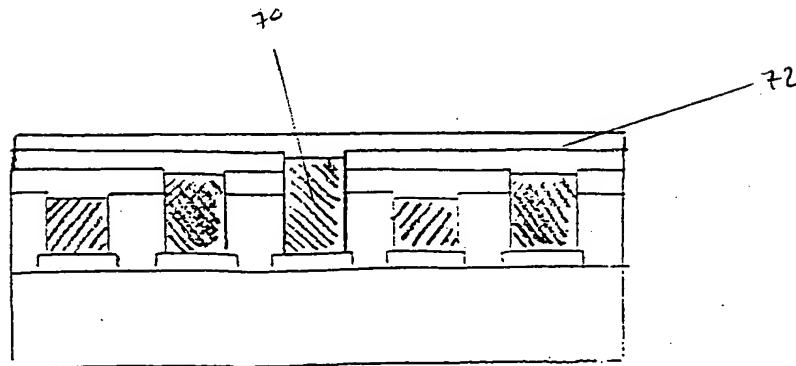


Fig. 2(h)